Gel Reaction Flow Patterns in a Non-Element Reactor with Two Branch Flows

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This master thesis would not have been possible without the generous help of my Sempai Masaki Yamaguchi, neither without the support of my family even if they were really far away from me…

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Abstract

In the framework of chemical and energy engineering fluids have been the subject of many studies for a lot years. In some cases, these fluids upon contact form gels. These gels have many applications like medical, food, cleaning... The peculiarity of these materials that share both properties of solids and liquids are yet to be described. In this study, a qualitative and quantitative analysis of the mass transfer is carried out when in a reactor without internal element 10 mass% PVA (polyvinyl alcohol) and 3 mass% Borax are introduced. The PVA solution was supplied to the main flow pipe at a constant inlet pressure of 20 kPa. The Borax solution was red dyed to visualize in a better way the flow pattern and it is introduced at a constant velocity of 30 mm/s during 16 seconds in a perpendicular direction to the non-element mixer. It has been previously studied that these compounds upon interaction form a gel. This gel, for its future industrialized production must be divided into independent capsules and for this PVA is introduced again by a new branch to the reactor in order to divide into two parts the formed gel pill. The second injection of PVA was dyed blue and parameters of velocity and chemical composition were varied in order to know which are the boundary conditions for the splitting phenomena. The analysis of the transfer of matter is carried out by a code (also developed in the present work) of Matlab of recognition of intensity of colors in images. The experiments have been recorded. For the quantitative analysis, a pressure sensor has been placed along the reactor which provides data along the reactor.

State of art

In the present state of the art it will be tried to disaggregate the knowledge and previous literature regarding this work. To this end, the division of the study has been considered in four parts, although different links between them exist. A first part where the particularities and characteristics of the gels will be analyzed, since in the present experiment it will be used a mixture of liquids (PVA and BORAX) that when they come into contact a gel film is formed. In the second part, it will be taken the subject of the mixers, referring to studies done in the past with similar devices and with the non-element mixer that is going to be used in this experiment. Regarding the third part, since it is intended to analyze the mass transfer rate it will be developed this concept and some previous work will be mentioned. For the analysis of this transfer phenomena a Matlab code will be used whose objective to analyze the color of the images taken by a photo-camera during the experiment. With these four sections is intended to give a general idea of the current state of knowledge referred to this topic.

Gels

Gels are positioning themselves in society as materials whose properties are innumerable. Its peculiar structure gives them a series of characteristics that distinguish them notably from solids or liquids. Because Brownian particles are electrically charged, there is interaction between them. This causes them to form a regular structure, which gives them a consistency that is not the rigid one of a crystal, but also the one of fluid that corresponds to the liquid. The common characteristic of the gels is the presence of a type of continuous structure which gives them the properties of

the semisolids. Certain gels have the ability to pass from one colloidal state to another, they remain fluid when agitated and solidify when they remain immobile. This feature is called thixotropy. Another property of the gels is the Weissenberg effect. Nearly all liquids have some degree of viscosity or elasticity, which is why, when you're stirring dough, a little will cling, but most will get thrown off. Only strongly viscoelastic fluids will be impressive when stirred by a rod. Gels are this type of fluids. Hence if a rotating rod is introduced inside a rheological liquid, the liquid instead of being displaced out of the rotor by the centrifugal forces towards the walls of the vessel, it starts to climb around the rotating rod. If instead of one rod, two coaxial cylinders are inserted, the rotation of the outer cylinder forces the liquid into the inner cylinder. This is a direct consequence of the normal stress that acts like a hoop stress around the rod.

The process by which a gel is formed is called gelation. Gelation processes are principally used in medical care, water treatments, food processing, material protection and adhesives among others.

Gels are materials where polymer chains form the links of a network immersed in a typically liquid environment. The polymer chains are cross-linked at the microscopic level by chemical bonds or weaker physical bonds; the type of bond is used to label the macroscopic material as a chemical or a physical gel, respectively. The physical bonds can have diverse origins, such as van der Waals interactions or hydrogen bonding, and can involve a complex local structure such as the formation of a small crystalline domain. Because of their significant liquid content (up to 99% liquid by weight), often comparable to conditions in physiological tissue, gels have found various applications, especially in biomedical contexts [1].

The different microscopic behaviors of cross-links in chemical gels and physical gels endow them with distinct macroscopic properties. Since the chemical cross-links prevent the chemical gels from dissolving in its

environment solvent, chemical gels behave macroscopically like solids. However, because of the weaker nature of the cross- linking bonds, physical cross-links are found in a constant cycle of creation and dissolution in physical gels. At short time scales, against quick deformation, the cross- links do not have time to dissolve and the gel shares the same solid-like behavior of chemical gels. At long time scales, bond destruction is able to eventually release all shear or anisotropic stress. In other words, physical gels can adapt to the presence of boundaries in much a similar way as a liquid at long time scales [1]. In this paper, we will use a mixture that generates the formation of a physical gel to analyze its properties in terms of mass transfer due to the hydrogen bonding.

A hydrogen bond exists when a hydrogen atom is bonded to two or more other atoms, a donor atom X and an acceptor atom Y. Since the hydrogen atom has only one orbital (1s) at sufficiently low energy, hydrogen bonds are mainly electrostatic in nature but covalent and repulsive orbital-orbital interactions are also present [2]. Depending on the type of X and Y, there are strong and weak hydrogen bonds. In the case of weak and very weak hydrogen bonds, hydrogen bonding is mainly electrostatic in nature. In the case of strong and very strong hydrogen bonds, covalent bonding phenomena are also of some importance.

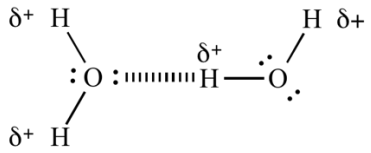


Fig. 1. Hydrogen bonding scheme

The hydrogen is attached directly to one of the most electronegative elements, causing the hydrogen to acquire a significant amount of positive charge. Each of the elements to which the hydrogen is attached is not only significantly negative, but also has at least one "active" lone pair. Lone pairs at the second level have the electrons contained in a relatively small volume

of space which therefore has a high density of negative charge while lone pairs at higher levels are more diffuse and not so attractive to positive things.

Micromixer

When mixing two liquids (multiphase reaction) an improvement of the mixture is generally achieved if it is carried out under turbulent regime. To do this, a static mixer is used, which is a reactor that has an internal element that enhances the contact surface between the two liquids for an optimization of the mixture. Hobbs and Muzzio (1997) [3] investigated the mixing process in a Kenics mixer experimentally, numerically and graphically. There are also studies by You et al. (2009) [4] that evaluate the state of the mix according to the mixer elements introduced. Because of the importance of mixing, a static mixing process is useful for in situ gelation. A static mixer is a tubular motionless mixer, inside which mixing elements can be inserted. When injecting fluids into a static mixer, it promotes the mixing of fluids by generating flows in the radial and tangential directions via these mixing elements. Depending on the geometry of the mixing elements, static mixers are categorized into several types, including the Kenics, SMX, or SMV types.

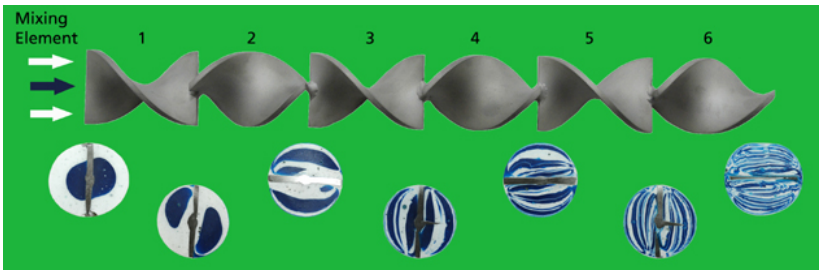


Fig. 2. Kenics Static mixer flow patterns [5]

However, the use of elements throughout the mixer can lead to a number of undesired effects. These effects encompass pressure losses due to

shear stresses in the tube walls when the fluid crosses the element and also limitations in size. That is why micromixers are less likely to be used with an internal element. On the other hand, when the two liquids that are mixed generate a gel capsule the fact of having an element inserted in between can difficult the formation of the capsule and its analysis. That is why in this type of experiments is recommended the use of mixers without internal element (non-element mixers). In addition, with this type of devices it is easier to analyze the flow pattern and the cleaning is more effective. In the fig. 3 it is possible to observe the internal shape of a non-element mixer [5].

Micromixers based on various principles have been produced and widely investigated. They can be classified into two main categories active and passive. Each of these micromixers has different capacity, operating requirements and mixing speed. For instance, an active micromixer requires power input in order to effect mixing in the device while a passive micromixer achieves mixing with the applied pressure for fluid motion. As such, depending on the application some micromixers are more suitable than the others [6].

For more than two decades, many efforts have been paid to apply microreaction technology to intensify multiphase reactions. The diameter of these microchannel is typically in the order of millimeter or less. One of the major applications of microreaction technology is multiphase reaction and it is because multiphase flow in microchannel exhibits characteristics flow pattern, that leads to an improvement of interfacial mass transfer rate [7]. In recent times, microchannel reactors have become a new and promising technology in chemical engineering, biotechnology and other sectors such energetic engineering. Processes based on multiphase reactions in microchemical systems, especially liquid-liquid two-phase reactions, occur in a broad range of application areas, such as nitration, extraction, emulsification, and so forth, and can form the basis for manufacture of a large variety of intermediate and consumer end-products [8].

In this area a large number of publications can also be found. Kashid et al. (2011) [9] analyzed the influence of the channel shape and the strong affection of the flow regime by using five different types of microchannels, which were T-trapezoidal, T-square, Y-rectangular, caterpillar and concentric. In this experiment six different flow patterns including annular and slug flows were observed.

Mass Transfer

When a system contains two or more components whose concentrations vary from point to point, there is a natural tendency for mass to be transferred, minimizing the concentration differences within the system. The transport of one constituent from a region of higher concentration to that of a lower concentration is called mass transfer. The mechanism of mass transfer, as we have also observed in heat transfer, depends upon the dynamics of the system in which it occurs. Mass can be transferred by random molecular motion in quiescent fluids, or it can be transferred from a surface into a moving fluid, aided by the dynamic characteristics of the flow [10]. On one hand in a multi-phase system mass is transferred due to the chemical potential differences between the species. On the other hand, a single-phase system where temperature and pressure are uniform, the difference in chemical potential is due to the variation in concentration of each species.

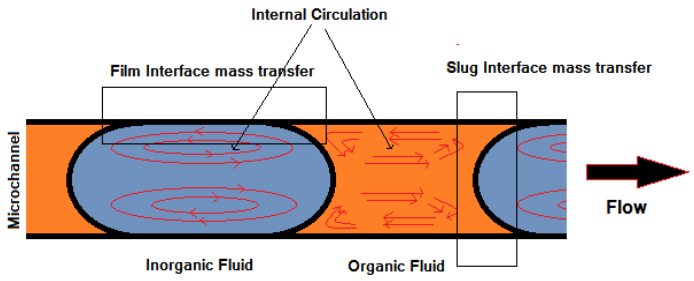


Fig 3. Scheme of boundary mass transfer zones [14]

The mass transfer rate and the flow pattern in microchannel are influenced by several factors. Among these factors, we have the velocity of the main flow and branch flow, the diameter of the microchannel and fluid properties. Also, the geometry, the shape of the cross section and contacting junction are factors that affect the flow pattern. To analyze these effects many papers and books have been written and they contain valuable information on the past work from the point of view of transport phenomena of multiphase flow in microchannels. The effects of flow condition on the flow regime in microchannel was summarized in detail by Kreutzer et al. (2005), Shao et al. (2009) [11], Rebrov (2010) [12] and fluid behavior by Gupta et al. (2010) [13] and Sobieszuk et al. (2012) [14].

Yuchao Zhao et. al (2007) [6] examined mass transfer characteristics of water, n-butanol and succinic acid with both counter and cross-flow T-junction micro-channels proposing correlations to predict the volumetric mass transfer coefficients. These coefficients resulted to be two or three orders of magnitude higher than those of conventional liquid-liquid contactors [6].

Yashihito Okubo et al. (2008) have investigated three extraction operations, contact flow in a Y-shaped micro channel, segmented flow and emulsification. In this paper, they notice that extraction using segmented flow is dependent on mass transfer by molecular diffusion. Hence its rate is enhanced by the internal circulation flow generated inside slugs and extraction rate can be controlled precisely by adjusting operational parameters such as the flow rate and the flow rate ratio. They also described that enhancement of mass transfer between two phases and improvement of extraction efficiency are expected if features of micro-spaces are adopted for a liquid–liquid two-phase process [15].

Nobuaki Aoki et al (2011) have examined that to increase the maximum flow rate that enables slug flow formation in miniaturized channels, a gas-liquid –liquid slug flow was formed when gas phase slugs were put together in a liquid- liquid slug flow. Effects of channel size, void fraction (ratio of volumetric flow rate or gas phase to that of total flow rate), and volume ratio of aqueous phase to organic phase on flow regime were examined. Results indicate that the total liquid-phase flow rate that forms a stable gas−liquid−liquid slug flow increases up to 200 mL/min with air addition (void fraction > 0.1) in a poly(tetrafluoroethylene) (PTFE) tube (inner diameter 3 mm). The mass-transfer rate in the gas−liquid−liquid slug flow was also high because internal circulation flow is enhanced by increased flow rate. The mass-transfer coefficient was then correlated with the channel internal diameter and the flow velocity [16].

Akbar et al (2003) has experimented with gas-liquid two-phase flow regimes and their transitions in microchannels whose cross-sections (circular and near-circular) are reviewed and compared. It was shown that the available data are in good agreement for microchannels with hydraulic diameters close to 1 mm. With these data it hjas beed developed a simple Weber number-based flow regime map that divides the entire flow map into four zones: the first one is a surface tension dominated zone including bubbly and plug flow patterns; the second refers to an inertia dominated zone representing the annular flow regime; the third is a dispersed/churn flow zone; and finally a transition zone that consists of other intermittent flow patterns [17].

Usha Rao (2012) has studied how the flow pattern mapping and dimensionless analysis of two phase liquid-liquid affect systems in glass micro channels of sizes that vary from 500 pm to 1000um and varying shapes such as straight and sinusoidal [18].

Image analysis

A simple and at the same time accurate way to visualize the flow patter is making use of Matlab. When two fluids come into contact in a microchannel there is a mass transfer that can be measured in terms of color variation. For this a Matlab code can be used. In the present work, the images obtained from the mixture of the analyzed liquids by a code will be analyzed. This will allow the mass transfer rate in the process to be quantitatively known. For this, a code has been developed which will be shown below.

There are numerous papers and books that have used image processing for pattern recognition as well as for color analysis. Among them, Milad Fathi et al. (2011) whose work was focused on the use image analysis and artificial neural network to predict mass transfer and kinetics changes of osmotically dehydrated kiwifruit slices, also Katherin et al. (2006) investigated about color measurement in L * a * b * units from RGB digital images [18].

Dessimoz et al. (2008) examined the mass transfer rate by the color change of bromothymol blue in the aqueous phase. In this experiment flow patterns were obtained for Y- and T-junction of two micro-channels and they were monitored by a camera. Volumetric velocities of water and organic phase were varied between 1 and 6 ml/h. The mass transfer performances of two flow patterns were compared and the formation mechanism of parallel and slug flow was studied. The shape of the interface between the immiscible liquids was controlled by a competition between the viscous forces and the local interfacial tension [19].

Pressure Analysis

Many techniques have been developed for the measurement of pressure and vacuum. Instruments used to measure pressure are called pressure gauges or vacuum gauges.

A manometer could also be referring to a pressure measuring instrument, usually limited to measuring pressures near to atmospheric. The term manometer is often used to refer specifically to liquid column hydrostatic instruments.

A vacuum gauge is used to measure the pressure in a vacuum—which is further divided into two subcategories: high and low vacuum (and sometimes ultra-high vacuum). The applicable pressure range of many of the techniques used to measure vacuums have an overlap. Hence, by combining several different types of gauge, it is possible to measure system pressure continuously from 10 mbar down to 10−11 mbar.

Although no pressure is an absolute quantity, everyday pressure measurements, are usually made relative to ambient air pressure. In other cases measurements are made relative to a vacuum or to some other ad hoc reference. When distinguishing between these zero references, the following terms are used:

- Absolute pressure is zero referenced against a perfect vacuum, so it is equal to gauge pressure plus atmospheric pressure.
- Gauge pressure is zero referenced against ambient air pressure, so it is equal to absolute pressure minus atmospheric pressure. Negative signs are usually omitted.
- Differential pressure is the difference in pressure between two points.

The zero reference in use is usually implied by context, and these words are only added when clarification is needed. Differential pressures are commonly used in industrial process systems. Differential pressure gauges have two inlet ports, each connected to one of the volumes whose pressure is to be monitored. In effect, such a gauge performs the mathematical operation of subtraction through mechanical means, obviating the need for an operator

or control system to watch two separate gauges and determine the difference in readings. Moderate vacuum pressures are often ambiguous, as they may represent absolute pressure or gauge pressure without a negative sign. Atmospheric pressure is typically about 100 kPa at sea level, but is variable with altitude and weather. If the absolute pressure of a fluid stays constant, the gauge pressure of the same fluid will vary as atmospheric pressure changes. Some standard values of atmospheric pressure such as 101.325 kPa or 100 kPa have been defined, and some instruments use one of these standard values as a constant zero reference instead of the actual variable ambient air pressure. This impairs the accuracy of these instruments, especially when used at high altitudes.
Use of the atmosphere as reference is usually signified by a (g) after the pressure unit e.g. 30 psi g, which means that the pressure measured is the total pressure minus atmospheric pressure. There are two types of gauge reference pressure: vented gauge (vg) and sealed gauge (sg).

A vented gauge pressure transmitter for example allows the outside air pressure to be exposed to the negative side of the pressure sensing diaphragm, via a vented cable or a hole on the side of the device, so that it always measures the pressure referred to ambient barometric pressure. Thus a vented gauge reference pressure sensor should always read zero pressure when the process pressure connection is held open to the air.

A sealed gauge reference is very similar except that atmospheric pressure is sealed on the negative side of the diaphragm. This is usually adopted on high pressure ranges such as hydraulics where atmospheric pressure changes will have a negligible effect on the accuracy of the reading, so venting is not necessary. This also allows some manufacturers to provide secondary pressure containment as an extra precaution for pressure equipment safety if the burst pressure of the primary pressure sensing diaphragm is exceeded.

There is another way of creating a sealed gauge reference and this is to seal a high vacuum on the reverse side of the sensing diaphragm. Then the output signal is offset so the pressure sensor reads close to zero when measuring atmospheric pressure.

A sealed gauge reference pressure transducer will never read exactly zero because atmospheric pressure is always changing and the reference in this case is fixed at 1 bar.

An absolute pressure measurement is one that is referred to absolute vacuum. The best example of an absolute referenced pressure is atmospheric or barometric pressure.

To produce an absolute pressure sensor the manufacturer will seal a high vacuum behind the sensing diaphragm. If the process pressure connection of an absolute pressure transmitter is open to the air, it will read the actual barometric pressure.

Experimentation

Working fluids

As discussed earlier in this study, two fluids will be mixed which when contacted will produce a gelling reaction. The working fluids are Polyvinyl Alcohol (PVA) and an aqueous solution of Borax. On one hand, Polyvinyl alcohol (PVOH) is a hydrophilic linear polymer that forms copolymers of vinyl alcohol and vinyl acetate. Therefore, the structural properties of polyvinyl alcohol polymers depend on the extent of polymerization and hydrolysis. Chemical and physical modifications are caused by changes such as esterification, etherification, crystallization, ion-polymer complexation in the polymer. Modified- PVOH structures are suitable in biomedical applications. It has the idealized formula $[CH_2CH(OH)]_n$. On the other hand, Borax ($Na_2B_4O_7 \cdot 10H_2O$, sodium borate or sodium tetraborate) is an important compound of boron. It is the commercial name of the boron salt. It is a white and soft crystal that dissolves easily in water and glycerol. Figure 4 shows the gel reaction scheme when aqueous solution of Borax and PVA are mixed. The crosslinking agent of the reaction is the Borax solution and the gel is formed with the aqueous solution of PVA.

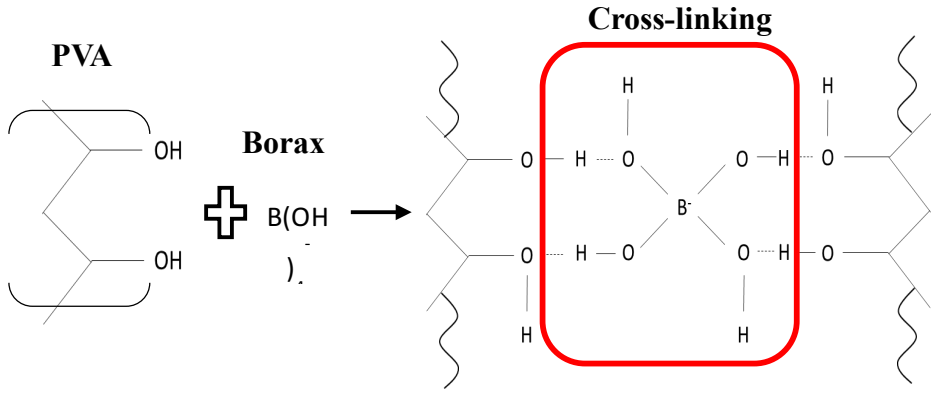


Fig 4. Gel reaction between PVA and Borax

In this experiment, we use 10 mass% PVA for the main flow and 3 mass% Borax for the branch flow. Since we know the conditions of formation of the gel capsule (studied by Masaki Yamaguchi et al. (2016), to achieve its division it will be used a new branch injection of PVA to analyze the flow patterns produced by dividing the capsule. This injection of PVA will have as variable the percentage of PVA in aqueous solution. This percentage will be varied from 10 mass% to distilled water going through all pair numbers (8 mass%, 6 mass%, 4 mass%, 2 mass%). With this segmentation of the mass percentage of PVA, the boundary conditions of division of the capsule will also be found. This is interesting because finding a limit value improves the cost structure for possible future applications. Properties of the working fluids (density, viscosity and pH) can be visualized in the following table.

Working Fluid	Viscosity [mPa·s]	Density [kg/m^3]
10 mass%PVA solution	33.3	1001
8 mass%PVA solution	21	1001
6 mass%PVA solution	12.4	1001
4 mass%PVA solution	7.44	1001
2 mass%PVA solution	2.42	1001
3 mass% borax solution	1.31	1000

Table 1. Properties of the working fluids

These values of viscosity have been taken with the aid of a rotating viscometer while the pH values have been realized with a ph-meter using glass electrode method. A magnetic mixer (Fine Magnetic Stirrer F-205) has been used for the preparation of the solutions. A magnetic stirrer or magnetic mixer is a laboratory device that employs a rotating magnetic field to cause a stir bar (also called "flea") immersed in a liquid to spin, thus stirring it. For the calculation of the mass percentage a scale has been used with precision to the hundredth of gram.

Experimental Apparatus

In this section, the equipment used for the experiment will be explained in detail. For this study only one equipment was used with which all experiments were performed. Previously, this equipment had been used by Masaki Yamaguchi et al. (2016) to determine flow characteristics with gel reaction in a non-element mixer. In their project, the parameters regulating the formation of the gel capsule were elucidated. Thus, a main flow and a single branch flow were used. In the present study, based on the work of Masaki Yamaguchi and collaborating with him, we try to divide the gel capsule into two equal parts. For this purpose, the introduction of a new branch flow (B2) after the entry of borax (B1) has been devised.

The equipment is constituted by a tank in which the 10 mass % PVA solution is stored. This tank is hermetically closed with the aid of 8 screws and during the realization of the experiment it is not opened at any moment. The liquid present in the tank is driven by a slightly pressurized Nitrogen reservoir at a pressure of P = 20 kPa (constrained to capsule formation). Nitrogen gas is an industrial gas produced by the fractional distillation of liquid air, or by mechanical means using gaseous air (pressurized reverse osmosis membrane or pressure swing adsorption). Nitrogen gas generators using membranes or pressure swing adsorption (PSA) are typically more cost and energy efficient than bulk delivered nitrogen. When supplied compressed in cylinders it is often called OFN (oxygen-free nitrogen).

Therefore, the PVA is introduced into the mixer at a preset pressure. Between the mixer and the supply tank an electric valve and a pressure detector are located. To calculate the Reynolds number in the main flow the following expression is used:

$$\mathrm{Re} = \frac{\rho_{pva} \cdot v_d}{\mu_{pva}}$$

Where ρ_{pva} represents the density of PVA 10 % mass solution and μ_{pva} the viscosity of the same solution. V is the average outlet velocity of the main flow and d is the inner diameter of the main pipe.

Fig. 5. Gas Tank for the main flow

The pressure sensor is connected to an A/D (analog-to-digital) converter whose function is to transform the analog pressure values by sampling in a digital signal. These transformed data can be interpreted by a PC and are displayed in a pressure graphic on an excel sheet. Immediately after the pressure sensor there is the mixer. As mentioned above, the mixer is a non-element mixer and therefore is a hollow pipe. The dimensions of the mixer are: an internal diameter of d = 10 mm and an external diameter of D = 12 mm and a specific length of L = 520 mm. This tube is perforated radially three times by 100 mm distance from the pressure sensor as is showed in the figure 5.

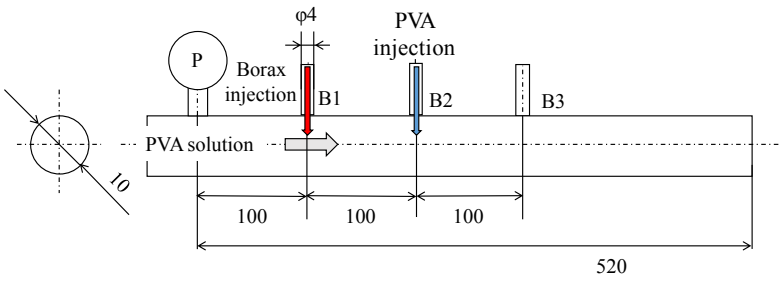


Fig. 6. Section of the non-element mixer

In these holes the branch flow is placed. In the first one the entrance of Borax takes place, which will form a gel capsule. In the second branch flow the PVA is injected by varying the concentration parameters previously described and the third perforation will serve as an instrument to extract the possible bubbles by creating a vacuum with a syringe. For each branch flow the relevant liquids will be injected using syringes connected to an electronic device that will measure the injection parameters. This device is a sequencer that uses a mathematical program to control the times of injection and the velocity injection. For the interpretation of the results, the whole procedure is filmed with a video camera. The following figure shows an outline of the switchgear used.

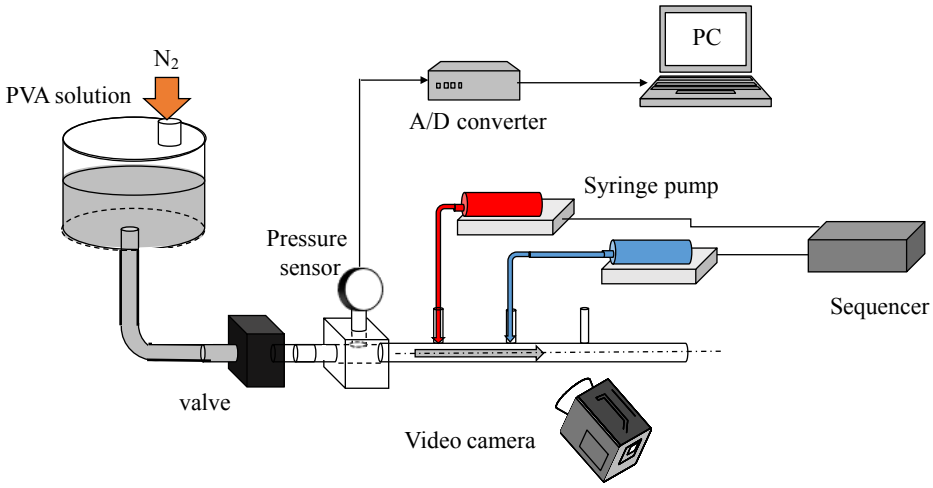


Fig. 7. Experimental apparatus schematic

As it can be seen in the figure dyes are used for each of the liquids used in order to facilitate their differentiation when image processing takes place. The borax solution is tinted red while the PVA solution injected in the branch flow is blue dyed. The PVA used in the main flow is colorless.

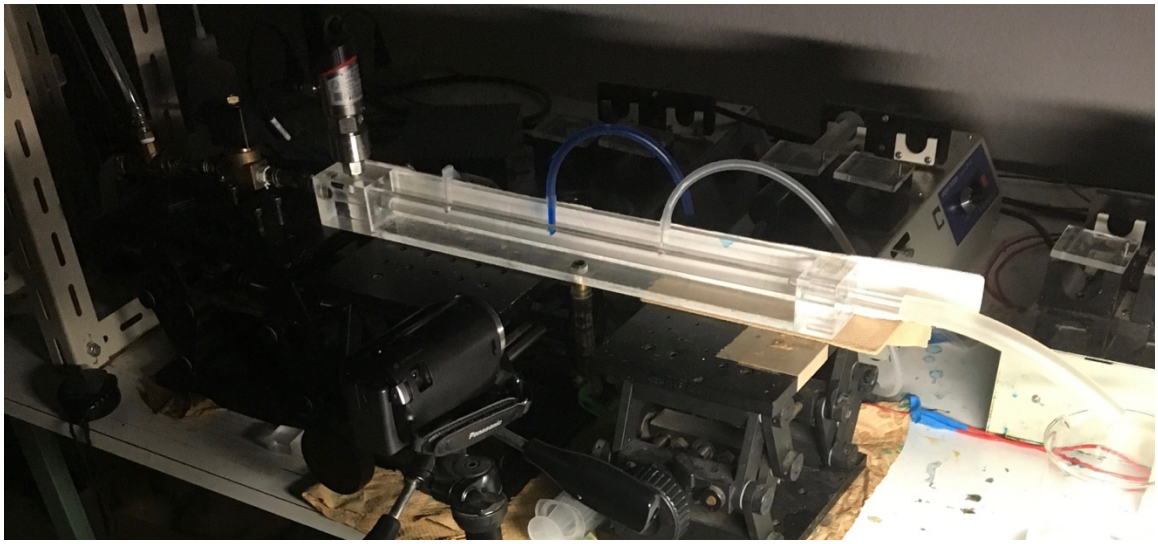


Fig. 8. Non-element mixer and camera

Methodology of the Experiment

This section describes the steps used to perform the project. Since this program has a part of experimentation and another part of theoretical analysis it has been considered to divide this module into two sections.

The first section focuses on the experimental part, done in the laboratory and it is divided again into two subsections. In the first of these subsections, the steps followed for a 10% mass PVA solution in which the injection velocity in B2 is varied is analyzed. In the second subsection, using the optimum speed for the separation of the capsule (obtained in the first step), the mass percentages of the PVA solution are varied in order to find the limit value for splitting the capsule.

The second section is dedicated to the programming part. As previously mentioned, for the analysis of the images taken with the camera has been considered the use of a mathematical code. Matlab has been chosen as the

more suitable program for this purpose. This part will explain in detail each step that is followed in the code.

PVA solution 10% mass varying the injection velocity

For the realization of this experiment, a main flow (parallel to the mixer) of a 10% mass PVA solution has been used as explained above. This main flow has been driven by the pressurized nitrogen tank at a pressure of 20 kPa. This pressure is the one previously studied by Masaki Yamaguchi et al (2016) for the formation of the capsule. It has been considered to approximate it so that the velocity of the PVA along the tube does not generate a Reynolds number greater than unity. In branch flow 1 (B1) borax dissolved in water to 3% mass has been introduced whose parameters of injection have been a velocity of injection V_{in} = 30 mm/s and a time injection T_{in} = 16 s. These parameters have remained constant during this experiment. In branch flow 2 (B2) a 10% mass PVA solution has been introduced as in the main flow.

The parameters varied in this experiment have been those of injection of PVA in the main branch flow. The variation of these parameters is shown in the table 2. Since it is intended to measure the transfer of mass it has been considered to inject in each of the tests the same amount of matter so that when increasing the rate of injection, the time of injection is decreased and vice versa.

V_{in} (mm/s)	0	15	30	60	120
T_{in} (s)	0	16	8	4	2

Table 2. PVA injection parameters in the B2

For greater precision in the data, it has been done three times for each speed and time of injection. In some cases, due to the appearance of bubbles or measurement errors it has had to perform extra times.

PVA solution varying the mass percentages

Once the previous experiment is finished and with the analyzed data, the present one is carried out. For the accomplishment of this experiment it is necessary to have previously determined what speed is the optimal one for the separation of the capsule (Borax-PVA gel reaction). Once determined, the percentage by mass of PVA is varied in order to determine the boundary condition for the division of the mixture. This will show us different flow patterns that will tell us which mass percentage is most appropriate for this purpose.

Mass Percentaje (%)	10	8	6	4	2	0
V_{in} (mm/s)	30	30	30	30	30	30
T_{in} (s)	8	8	8	8	8	8

Table 3. PVA injection parameters in the B2

In Table 3 we observe that the only variable parameter is the composition of the aqueous solution of PVA. In the same way, as in the previous experiment, it has been considered as adequate the repetition three times of each composition.

MATLAB code for color analysis

MATLAB is a high-performance language for technical computing. It integrates computation, visualization, and programming in an easy-to-use environment where problems and solutions are expressed in familiar mathematical notation. Although MATLAB is intended primarily for numerical computing, an optional toolbox uses the MuPAD symbolic engine, allowing access to symbolic computing abilities.

For the analysis of the images a series of steps are followed that are shown in the following phase diagram:

Flow Diagram 1. Code processes

Read the sample image

First of all, the image has to be read and for that *imread* function is used. *Imread (filename)* reads the image from the file specified by filename, inferring the format of the file from its contents. If filename is a multi-image file, then *imread* reads the first image in the file.

```
% Sample image
RGB = imread('C:\Users\ueda lab\imagexample.jpg');% File
name

% Separte RGB stack
R=RGB(:,:,1);
G=RGB(:,:,2);
B=RGB(:,:,3);
```

Then the indexed image is converted to RGB image. The RGB color model is an additive color model in which red, green and blue light are added together in various ways to reproduce a broad array of colors. The name of the model comes from the initials of the three additive primary colors, red, green and blue.

Read the base image

In this step the base image is read. This consists of an image of the reactor through which only the main stream of PVA to 10 percent by mass passes. To do this, the same function of the previous step is used. The colorimetric difference between the two images is then calculated to obtain the color map to be analyzed.

```
% Initial image
RGB1 = imread('C:\Users\ueda lab\imagexample.jpg'); % File
name
% Separte RGB stack
R1=RGB1(:,:,1);
G1=RGB1(:,:,2);
B1=RGB1(:,:,3);
% (Initial image)-(Image which we want to analize)
dR=abs(R1-R);
dG=abs(G1-G);
dB=abs(B1-B);
```

Color intensity Map

This process shows the color map of the image. It shows the intensity of both the red and blue color and for this the following functions are used.

```
% show mesh image;
mesh(dR)
 view([1 1 10])
 figure
mesh(dB)
view([1 1 10])
```

The function *mesh(Z)* draws a wireframe mesh using $X = 1:n$ and $Y = 1:m$, where *[m,n] = size(Z).* The height, *Z*, is a single-valued function defined over a rectangular grid. Color is proportional to surface height. The values in *Z* can be numeric, date-time, duration, or categorical values.

View([x,y,z]) sets the view direction to the Cartesian coordinates x, y, and z. The magnitude of *(x,y,z)* is ignored and figure creates a new figure window using default property values. The resulting figure is the current figure.

Analyze test section

Once the color intensity map of the entire image is obtained, it is interesting to stick to the area to be analyzed. To do this we must limit a test section. Figure 10 shows the test section to be examined. In order to generate a graph such as the figure 10, the average color intensity in the cross section is calculated. For it is made use of the mean function.

```
 % Test section
dR_test=dR(717:837,:);
figure
dB_test=dB(717:837,:);
mean_dR_test=mean(dR_test);
mean_dB_test=mean(dB_test);
```

```
plot(mean_dR_test)
xlabel('pixel');
ylabel('Blue intensity')
figure
plot(mean_dB_test)
xlabel('pixel');
ylabel('Red intensity')
```

Extra section: Excel sheet

To synthesize the results in a more effective way have been added lines of code that serve for the automatic generation of an excel file.

```
% write excel file
Color='trial.xlsx'; % file name
sheet1=1;
sheet2=2;
xlswrite(Color,mean_dB_test,sheet1);
xlswrite(Color,mean_dR_test,sheet2);
```

A file is created with two independent sheets showing the color intensity maps of the cross section for both red (Borax) and blue (PVA). *Xlswrite (filename,A,sheet)* is a function that writes to the specified worksheet.

Limitations of the Study

Although the results obtained are interesting, this paragraph shares some of the limitations of the study. One of the limitations of this study lies in the approximation of the PVA mainstream velocity. This, as discussed above has been fixed in function to the previous studies developed by Masaki Yamaguchi *et al*. The condition of formation of the gel sheet, enunciated in the mentioned previous studies, is restricted to a number of Reynolds lower than the unit. Since this velocity is set with a pressure measurement through the Nitrogen tank, this is not always the same. The Nitrogen tank used has an analog pressure gauge of spherical shape with an accuracy of 0.01 MPa as seen in the image. The condition that gives us in a permanent regimen a Reynolds inferior to the unit has been set at 0.05 MPa.

Fig. 9. Section of the non-element mixer

Another limitation associated with the study is related to the quality of the images obtained. For the study of the transfer has been used this method, measurement of the intensity of the colors for both working fluids. As the stocks of both compounds were not unlimited in many cases have been recycled. This may lead to an increase in dirt and an accumulation of impurities which may vary the behavior within the reactor. In some cases,

experiments have had to be undone by the appearance of anomalies in the color intensities due to these impurities or to dirt inside the reactor. On the other hand is the resolution of the camera. The transfer of matter between both fluids can only be appreciated in the form of pixels, since it is the maximum resolution reached by the apparatus. It is true that when two fluids occur a transfer of matter this (according to the explained law of Flick) occurs at the molecular level. Since a molecule is smaller than the pixels in the scale to which this experiment is being recorded, we are faced with a limitation in the knowledge of mass transfer.

It is also interesting to note that, since this project is framed within a major one, namely the first phase of a doctoral thesis elaborated by Masaki Yamaguchi, there are unknowns that cannot yet be revealed. One is whether this mass transfer between both fluids occurs at the level of the pipe walls, or if the PVA particles are otherwise able to pass through the gel sheet formed at the interface. For the analysis of this phenomenon a subsequent procedure should be followed when the experiment has been carried out. Once the capsules are expelled from the reactor by the pushing effect of the PVA (main flow) the analysis of the same is complicated, because when leaving the tube they lose their initial shape caused by the effect of the pressure on the walls of the tube. Once both capsules have been extracted they should be analyzed via color recognition using images or with the aid of a colorimeter. But as discussed above, this analysis is beyond the scope of this study because of the unavailability of apparatus and time.

Results

Once explained the steps followed for the accomplishment of work the results obtained are mustered. This section will be divided into the same parts as the previous one. The results obtained will be explained from a qualitative and quantitative point of view.

PVA solution 10% mass varying the injection velocity

Two types of pattern flow have been observed when the injection rate of the 10 %mass PVA solution has been varied.

First, when we do not inject PVA (figure 10) we observe how the gel capsule is formed. In this case, there is a transfer of minimum matter in the main flow, because the surface tension is greater in the walls of the tube and the gel particles are adhered. The parameters of borax injection for this phase are shown in the methodology section.

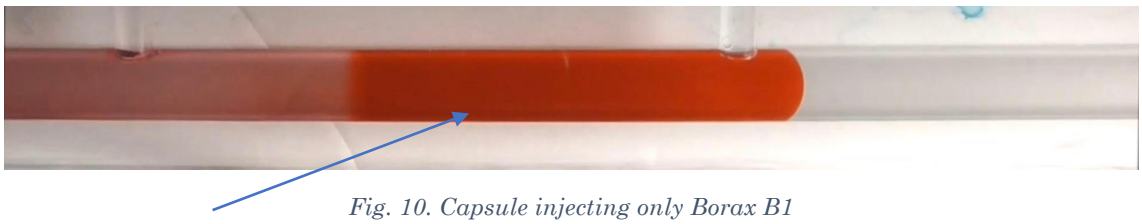


Fig. 10. Capsule injecting only Borax B1

When we injected 10% PVA in mass from B2 (Branch 2) we obtain different flow patterns for each treated velocity. It is evident that the division of the gel thru the PVA must be injected while the capsule is passing. In

Figure 18, the optimum moment at which the capsule is to be injected is precisely observed.

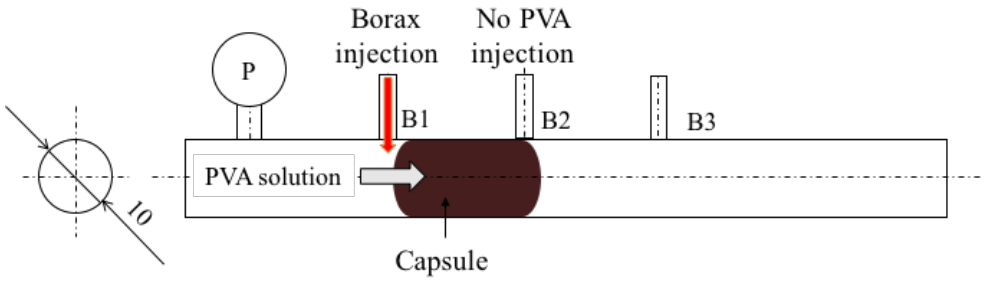


Fig. 11. Side-section of the non-element mixer

Image 12 shows the flow pattern for an injection of borax whose parameters are (V_{in} = 30 mm/s ; T_{in} = 16 s) and a posterior of PVA whose parameters are (V_{in} = 15 mm/s ; T_{in} = 16 s). As mentioned above, to maintain the amount of matter introduced into the experiment the speed and time of injection have been varied. This is a limitation in the study when we are injecting PVA 10% mass. The injection time from both branch flows is the same therefore the capsule does not have enough space to be divided. At the end of the injection from B2 the amount of Borax-PVA is scarce, thus the PVA agent cannot be introduced between the two fluids, occupying the upper part of the tube and not achieving a division.

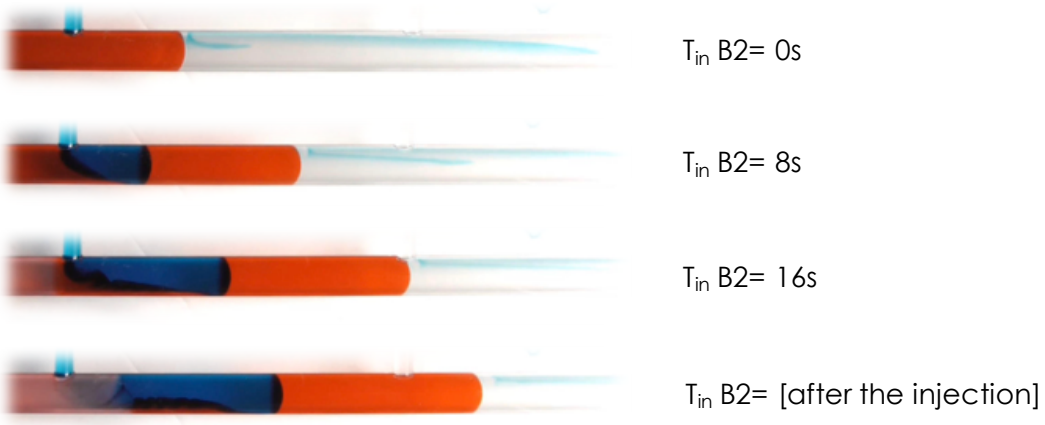


Fig. 12. Pattern flow PVA (V_{in} = 15 mm/s ; T_{in} = 16 s)

When, on the other hand, the injection parameters are the following (Borax (V_{in} = 30 mm/s ; T_{in} = 16 s) and PVA 10% mass (V_{in} = 30 mm/s ; T_{in} = 8 s)) we observe a different pattern flow. Because the time of injection of PVA is less than the formation time of the capsule, it is completely divided. It is experimentally observed that there is a mass transfer between the PVA island and the gel pill, which darks the posterior part of it. Through the image analysis program, a gap in color difference is observed, for both red and blue lines. This can be produced by the existence of a greater transfer of matter in the walls due to the velocity profile inside the tube. This profile shows a greater speed in the zones near the axis and smaller in the zones near the wall, approaching a parabola.

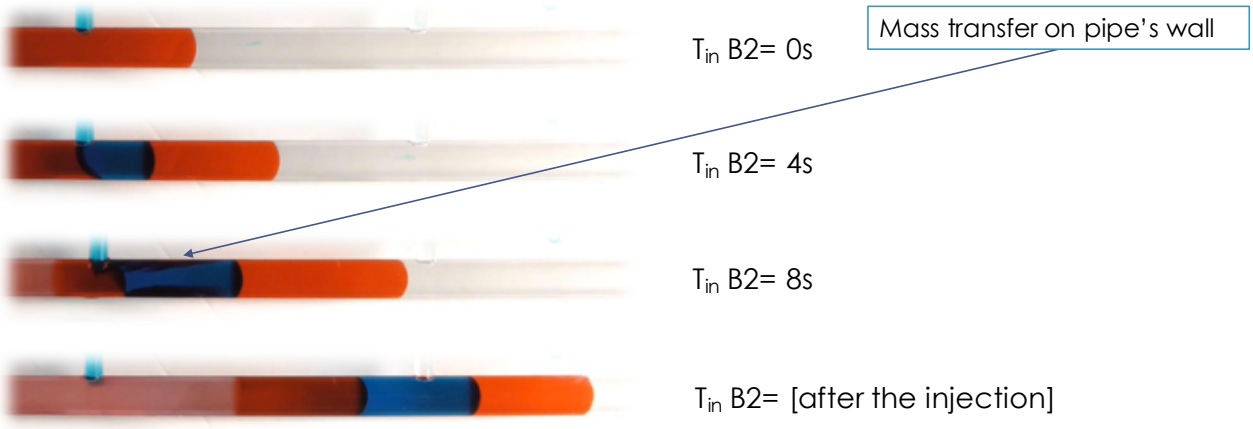


Fig. 13. Pattern flow PVA (V_{in} = 30 mm/s ; T_{in} = 8 s)

Something different happens when we increase the speed of injection of the PVA, maintaining the one of Borax, until V_{in} = 60 mm/s and T_{in} = 4 s. We can observe that at the time of injection the flow pattern is turbulent. Under these conditions the mass transfer is greater between the PVA island and the gel capsule. In the image provided by the Matlab code it is observed that the jump between the initial red and the one behind the capsule is larger than in the previous case.

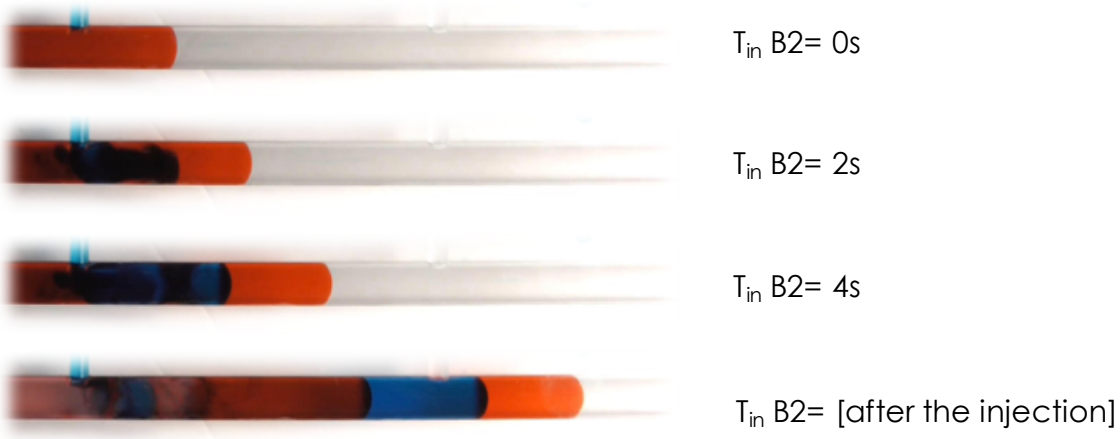


Fig. 14. Pattern flow PVA (V_{in} = 60 mm/s ; T_{in} = 4 s)

It is true that the capsule is separated, but the fact that at the time of injection the Reynold's number is increased in the branch flow (B2) causes a large part of the particles to flow randomly along the tube. This phenomenon causes some of them to move backwards and others forward, therefore the injection is not homogeneous and we find small islands of PVA along the capsule.

This same phenomenon happens when it is intended to be injected at higher speeds. In Figure 15 we observe the flow patterns when the injection and time ratios are V_{in} = 120 mm/s and T_{in} = 2 s respectively.

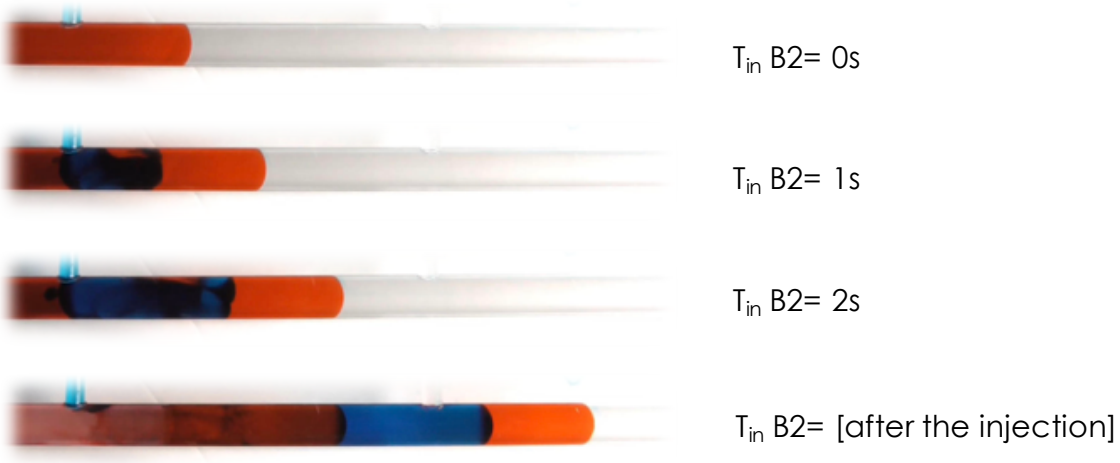


Fig. 15. Pattern flow PVA (V_{in} = 120 mm/s ; T_{in} = 2 s)

Once analyzed all the rates and times of injection it can be assumed that when injecting PVA under the following parameters V_{in} = 30 mm/s ; T_{in} = 16 s the capsule is divided into two halves evenly. This splitting is uniform. In addition, the gap shown by the color analysis is smaller, therefore there is a lower mass transfer between the PVA island and the gel formed. This is optimal in the case of industrial production since the less mass transfer between the interfaces the more robust the system will be for a possible application in the society (industry).

Pressure distribution

With the aid of the pressure gauge pressure data has been obtained along the tube. In graph 16 the average values of pressure are shown for each of the experiments. Since each experiment (same speed) has been performed three times, the mean has been calculated from those three times.

Given the differences between the pressure lines between the cases in which the capsule is divided and those not, it has been considered to place the graphs separately. In the first one it is observed how the behavior of the pressure has a similar profile for each velocity. There is no direct relationship that establishes that at a higher speed a pressure increase occurs in the reactor. The maximum pressure in all three cases is similar and takes a value of 0,43 kPa.

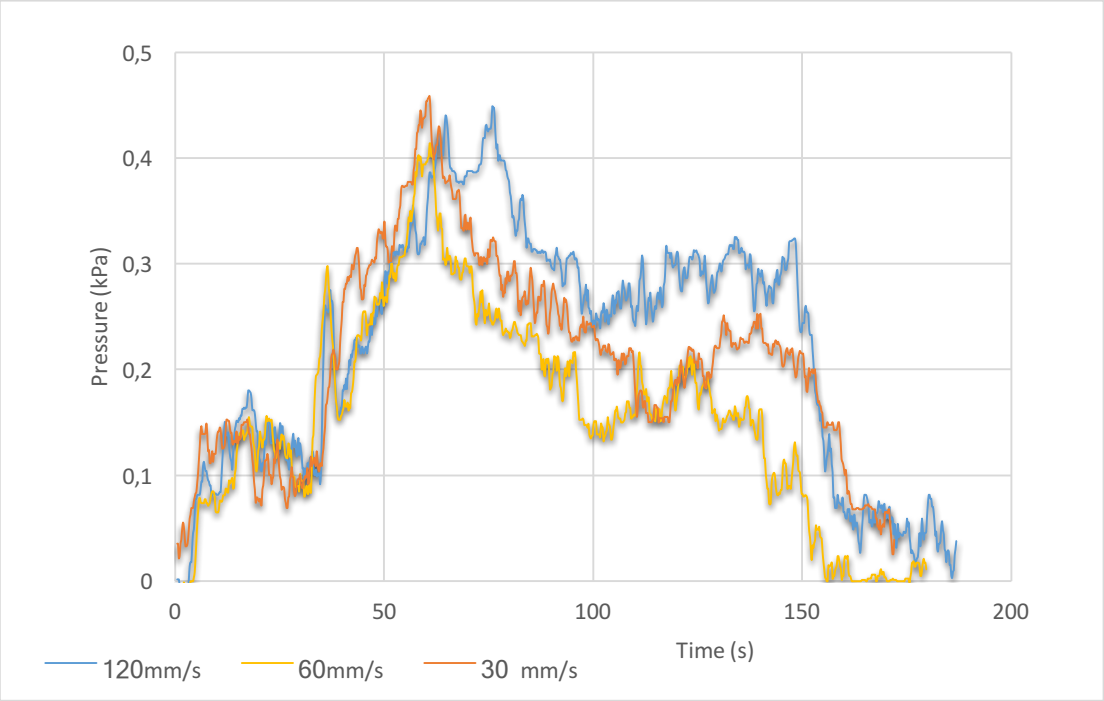


Fig. 16. Pressure distribution along the cross-section

On the other hand, in the following graph, the variation of the pressure is shown for a speed of 15 mm/s. In this case, explained above, the division phenomenon does not occur. Similarities are found between this case and when PVA is not introduced by the two input. The main difference is that there is an increase in pressure when PVA is introduced into the system corresponding to t = 60 s. At this point the pressure peak reaches a value of 0,27 kPa. In the case of Borax only injected by the first branch the pressure is maintained practically constant with decreasing tendency, whose maximum is 0,12 kPa which corresponds, as well as the grey line, with the moment of injection of Borax into the system.

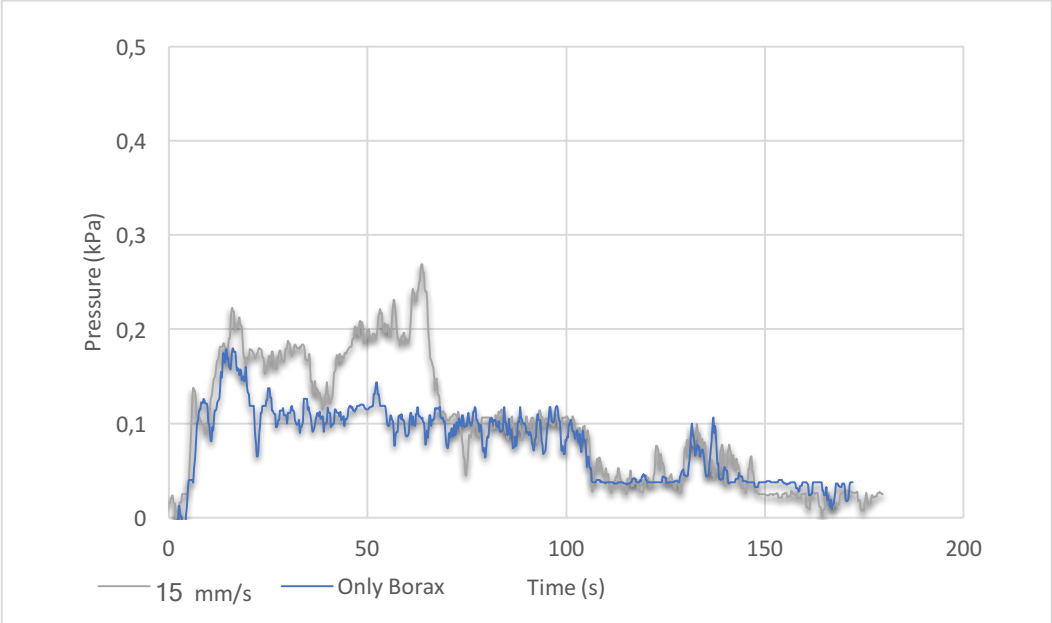


Fig. 17. Pressure distribution along the cross-section when no split.

It is concluded, in view of the results, that there is no direct relation between pressure and velocity, since there is no pattern of behavior in the figure 17. But there is a relationship between the pressure and the observed phenomenon. When the phenomenon of division of the gel pellet occurs the pressure follows a similar behavior and is different from that which follows when no such phenomenon occurs.

PVA solution varying the mass percentages

For this section, the optimum injection conditions analyzed in the previous section are used. This means that all the experiments performed in this phase fulfill the following parameters. On the one hand the Borax is injected at an inlet speed of 30 mm/s for a time of 16 s. On the other hand, the PVA (whose mass percentage of dissolution is varied) is injected at an injection rate of 30 mm/s for 8 seconds.

From the above experiment, it is known that if PVA 10% mass is introduced instants before half the gel film has passed with a velocity of 30 mm/s during a time of 8 s, it is separated. It has also been observed that this separation is uniform and minimizes mass transfer over other speeds used. Figure 18 shows the flow pattern of the mixture when using PVA 10% mass with these parameters.

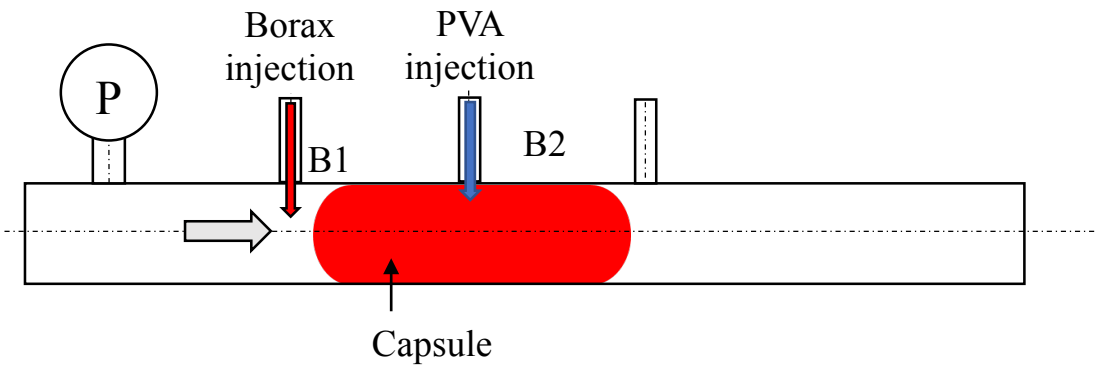


Fig. 18.Capsule formation

The injection process is the same as in the previous experiment. In this case it is interesting to analyze from the point of view of colorimetry the mass transfer existing when the percentage of PVA in the aqueous solution is varied.

Flow Patterns Observed

Two types of flow pattern were observed during the performance of this experiment attending to the mass percentage of PVA. Splitting pattern and layer pattern. Between them it was observed a different pattern that has been considered to be called transition

Splitting Pattern

This distribution of the gel and PVA masses was observed with 10, 8 and 6 mass% of PVA. In this case it can be observed, as well as in the previous experiment, that between the two gel capsules is located an island of PVA (Image 19).

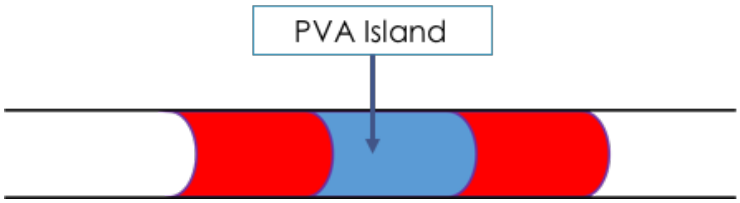


Fig. 19.Splitting Pattern flow

Within this pattern of division of the gel sheet differences have been observed when the percentages have been varied. As can be seen in figure 20 the posterior capsule color varies depending on the amount of introduced mass of PVA. Thus, the greater the percentage of mass introduced, the greater the intensity of the red color of the posterior capsule. This light / dark indicator is a useful way to measure mass transfer in the system. The fact that the red intensity of the rear capsule is smaller in each case (decreasing mass percentage) is an indicator that when we decrease the

amount of PVA introduced in the system there is more matter transfer between the PVA island and Gel sheet.

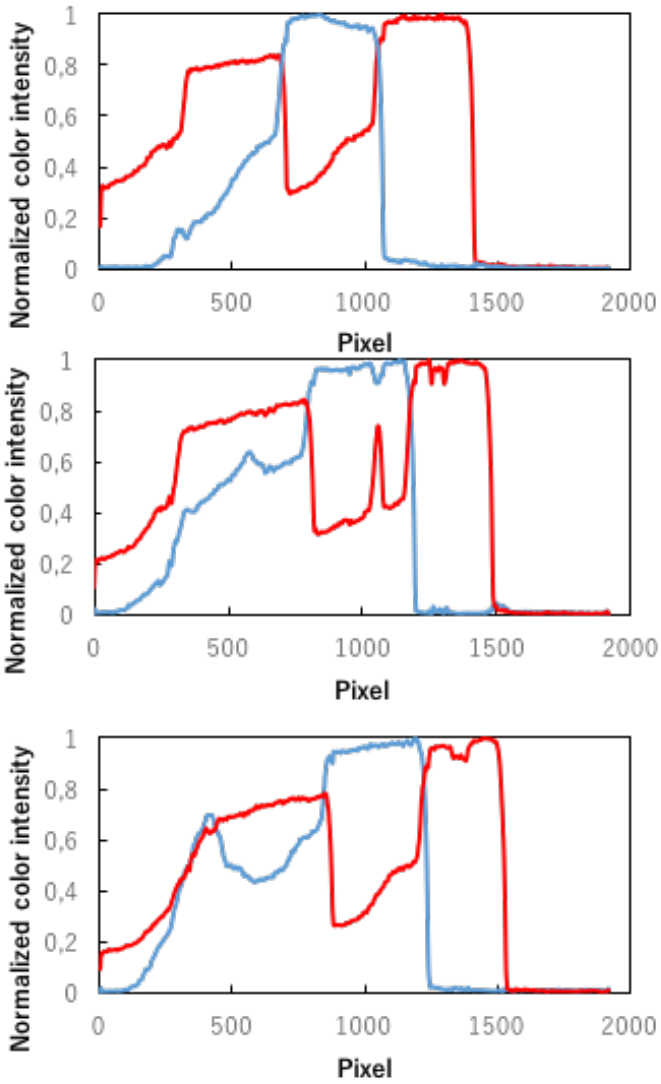


Fig. 20. Splitting Pattern Color Analysis. From up to down, 10, 8 and 6% mass PVA

For a better interpretation of the results it has been considered to include the real images that correspond to the color diagrams. The order is from top to bottom 10, 8 and 6% by mass, in the same way in color charts. In all the figures, it is observed that such a division of capsules exists. In the case of the second image, just as in the diagram it is observed how the island of PVA seems to be divided into two parts. This is an experimental error, since the injection of PVA may not be continuous due to a sequencer failure. The

consequences in this case are a minimum gel capsule (Borax-PVA) formed between the two islands of PVA.

Fig. 21. Splitting Pattern Real Image. From up to down, 10, 8 and 6% mass PVA

Transition Pattern

When injected into branch two PVA 4% in mass the pattern observed is called transition pattern. As mentioned above each experiment has been repeated 3 times. In this case, it has been observed that in one of the three attempts the capsule has been divided and in the other two it has not. It is therefore the lower limit of PVA that we must introduce to achieve a division behavior. It has been deduced, based on the observed results, that when PVA is introduced in 4% by mass it is able to generate an island of PVA in the middle of the capsules once divided. This phenomenon only happens if no fluctuation or disturbance is introduced into the system. At the moment when there is an anomaly that causes a fluctuation of certain variables, as well as an increase in the speed of injection due to the tolerance of the instrumentation, or a change in the pressure from the main tank, this system will not be able to annihilating it and therefore it will feedback itself positively. When this happens a pattern flow is observed as in figure 22. On the other hand, when this fluctuation (due to experimental errors) is not

introduced or is small enough to be eliminated, the pattern flow observed will be as in Figure 22.

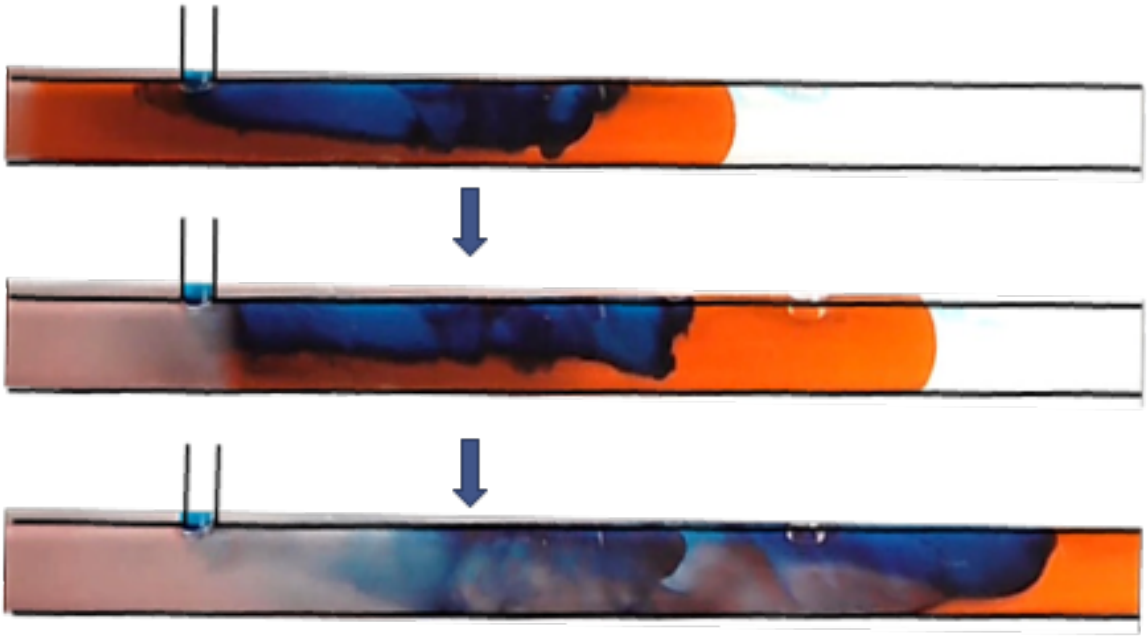


Fig. 22 (a). Transition Pattern Real Image. Non-split capsule

Fig. 22 (b). Transition Pattern Real Image. Split capsule

Layer Pattern

This phenomenon is observed when PVA 2% mass is introduced into the reactor. As seen on the table 1 the viscosity of the PVA solution decreases as its mass percentage decreases. In the case of 2% by mass this viscosity is fifteen times lower than that which it possesses with 10% by mass. When this fluid is introduced into the non-element mixer, its low viscosity has an effect on the action it generates on the gel. Viscosity is a measure of the resistance of a fluid which is deformed by either shear stress or tensile stress. In everyday terms (and for fluids only), viscosity is "thickness" or "internal

friction". When a low viscosity fluid collides with a much higher viscosity fluid, a number of forces appear at the interface of both. If this fluid, always injected at the same speed, varies its viscosity from highest to lowest, the forces at the interface will also vary from less to greater. That is why the force associated with the injection of PVA 2% mass is not high enough to divide the capsule and reach the lower zone of the reactor. In this case, what is observed in figure 23 happens.

Fig. 23. Layer Pattern Scheme Image.

There will then be two distinct layers, one of PVA and one of gel (Borax-PVA) that are distributed in parallel along the horizontal axis. The following figure shows this behavior taken from a photograph of the experiment.

Fig. 24. Layer Pattern Scheme Image.

Since the introduced PVA is not able to reach the bottom of the reactor for the above explained reasons, the section through which it circulates will be smaller than that of the tube. As the flow introduced by the second input (branch 2) is constant for the whole experiment, it is preserved. This causes an increase in the speed of diffusion of the PVA. The speed of the blue liquid will be higher than that of the lower gel capsule. When this situation occurs the collapse of the gel capsule occurs in the front. The following images illustrate this phenomenon.

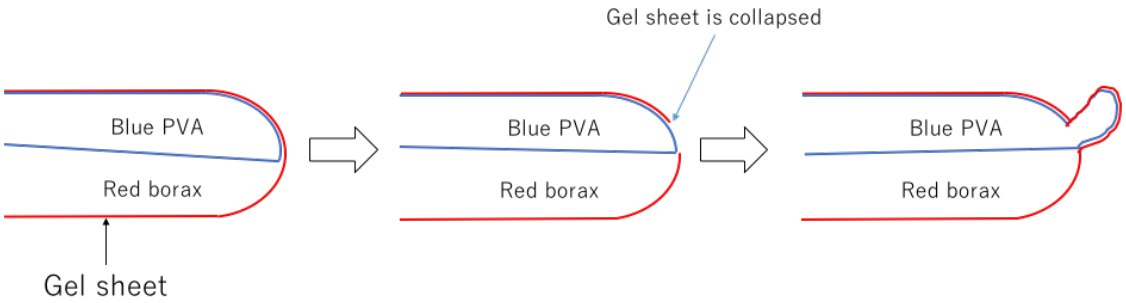


Fig. 25. Collapse of gel sheet Scheme Image.

Fig. 26. Collapse of gel sheet Real Experiment Image.

The analysis by means of the Matlab numerical control program clearly provides what is observed experimentally. The two colors maintain practically their intensities and there is not a balance (red appears/ blue disappears) like the one observed in the cases in which the gel divides in two.

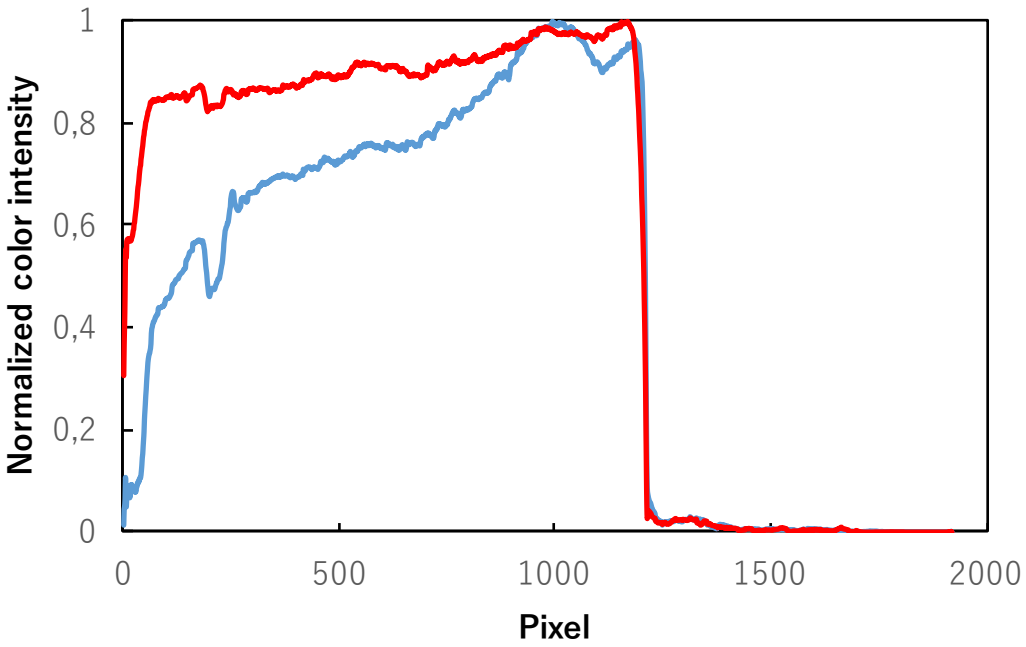
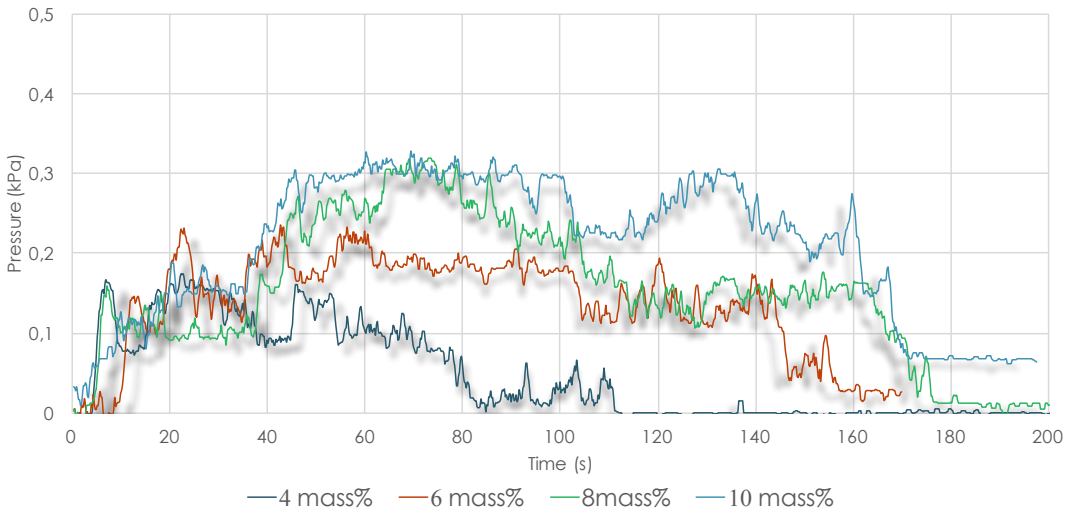


Fig. 27. Color analysis of the Layer Pattern

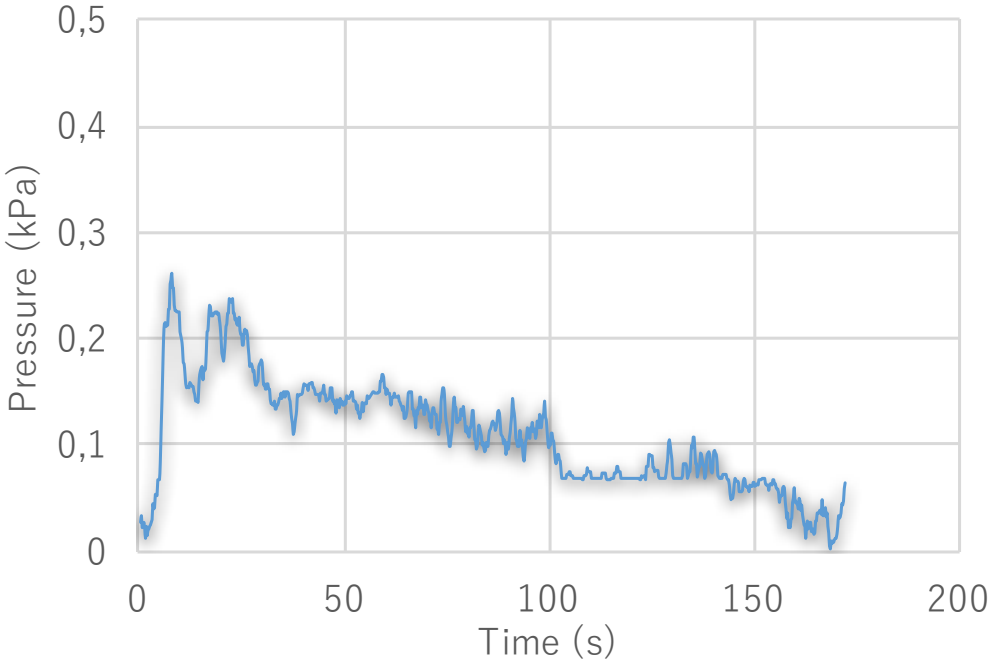
Pressure Distribution

Pressure measurement is always an important fact in an experiment inside a reactor. It can provide us with patterns that are established when we vary variables. The objective of the pressure analysis in this paper is to find a pressure concordance when the same results are obtained. With this we find that there is a similarity when the capsule is divided. The higher the mass percentage of PVA introduced into the system the greater the internal pressure line of the reactor. On the other hand, when this percentage is not high enough to divide the capsule the pattern of pressure found is similar to that of injection only of Borax by the first entry (branch 1).



Graph. 3. Pressure variation when %mass is varied

In graph 3 a large difference between the pressure curves of 10, 8 and 6% PVA is observed respect to that of 4% PVA. This 4% curve corresponds to the tests in which a complete division of the gel sheet was not achieved. While graph 4 (lower) shows the pressure curve when only Borax is introduced. Comparing both curves shows the same behavior at pressure level. The only difference between these two pressure lines is the increase produced by the injection of PVA at a given moment (shown in the graph 4).



Graph. 4. Pressure variation when only borax is injected

Conclusions

In the present experimental study, it has been tried to study and analyze the physico-chemical processes existing in the gelation process that occurs when PVA and Borax are put in contact. For this, and as explained above, several experiments have been performed.

The main flow was 10% in mass PVA solution driven by a steady input pressure, 3% in mass of Borax solution periodically injected from a side branch and variable conditions of injection of PVA. Referring to PVA, both the injection rate and the composition of the aqueous solution have been varied. The Reynolds number of the mainstream has been kept constant and less than unity since it has previously been studied that it is a necessary condition for the formation of the initial gel capsule when 3% borax by mass is introduced to the mixer without element. A pressure sensor has been placed at the inlet of the reactor whose purpose is to measure the pressure along the tube in order to find some type of pattern.

When the injection rate and the time of the PVA 10 mass% are varied, different behavior patterns are observed inside the reactor. It has been observed that at an injection rate of $v = 15\ mm/s$ and a time of $16\ s$ there is no division into two halves of the capsule because the injection time is too high. This means that when the PVA injection ends, it does so at the same time that the gel pill has passed the B2. Therefore, there are not enough particles to form the gel in the back. When, on the other hand, PVA is introduced by B2 at a rate of $v = 30\ mm/s$ for a time of $8\ s$, the formation of the desired PVA island is produced and the two halves of the formed gel are placed on both sides of she. For higher velocities such as $v = 60$ and $v = 120\ mm/s$ a different matter transport phenomena occurs. When entering at high speed through a narrow passage ($d = 4\ mm$) and being in its passage with the gel capsule, the particles of the Polyvinyl Alcohol either go forward or

backward which causes a greater isolated interaction of particles of this with the zone of gel. In this case, there is a greater transfer of matter between the two fluids, which makes the colors of both (as shown in the diagram provided by Matlab) altered.

For a quantitative analysis of this experiment the pressure sensor has been used. This provides the pressure values along the pipe (side-section). It is concluded that in the distribution of the pressure there is no direct relation between the rate of injection (and its corresponding time of injection) and the pressure. Since the maximum pressure studied corresponds to a speed of $30\ mm/s$ and has a value of $P = 0.4575\ kPa$ after 60.6 seconds while the maximum pressure when injected at a speed of $120\ mm/s$ is $P = 0.4475\ kPa$. In all cases of very close values of pressure it has been considered to conclude that for this range of velocities the pressure is not altered. However, the pressure behaves differently when the division phenomenon occurs or not. We therefore find a similarity in the pressure lines between the values of *30, 60* and $120\ mm/s$ which is different from those obtained when only Borax is introduced into the reactor and when the velocity is 15 mm/s.

When the percentage of PVA in aqueous solution ranges from 10% to 2%, three types of reaction are observed between the gel and the introduced PVA. In this case, and as explained in the experimental procedure section, the one that has provided the best gel capsule division conditions ($v = 30\ mm/s\ t = 8\ s$) has been used as the injection parameter.

Three distinct mixing patterns are observed. The first, observed for values of composition of 10, 8 and 6 mass% of PVA has been called splitting pattern for its divisive card, the capsule is able to divide evenly between two portions of Borax-PVA (gel) an island of PVA. From the values obtained by the analysis of images by Matlab it is observed that the mass transfer between the PVA island and the gel is greater the smaller the percentage by mass of PVA. This is visualized in the colorimetric graphs of the figure 20. In

those graphs the normalized blue intensity of the posterior capsule increases as the percentage of the aqueous solution decreases and there is a decrease in the intensity of red color. This phenomenon, is produced by the difference of viscosity that exists between the different chemical compositions.

For values of 4% in mass it has been observed that both phenomena take place, since in function of the experimental conditions in some cases the mixture gets to be divided whereas for other cases this phenomenon does not take place. This pattern has been called transition pattern.

Finally, for a 2% mass value of PVA, a different geometry is achieved. Due to the differences in viscosity of the two fluids upon contact, a lamination of them occurs leaving on the top of the gel capsule the introduced PVA coming to occur a collapse due to the increase of speed of the liquid introduced by the B2 caused by the decrease of the section through which it circulates. In this type of cases, the higher fluid velocity, as it is higher (Bernoulli) reaches the interface between the PVA injected by the tank (main flow) and the gel capsule causing a collapse in the direction of the reactor shaft.

References

[1] Yonghao An, Francisco J. Solis, Hanqing Jiang, "A Thermodynamic Model of Physical Gels ", School for Engineering of Matter, Transport, and Energy, Arizona State University, Tempe, 2, 2007.

[2] Jamie Tuttle, "The Nature of the Hydrogen Bond and its Application Towards Enantioselective Catalytic Reactions" ,MacMillan Group, 2005.

[3] D.M. Hobbs, F.J. Muzzio, "Reynolds number effects on laminar mixing in the Kenics static mixer", Department of Chemical and Biochemical Engineering. Rutgers University,Piscataway,1997

[4] You, S.H.N. Higuchi and T.Ueda, "Macroscopic estimation Method of the Mixedness of Kenics Type Static Mixer", Korean Journal of Chemical Engineering, 26, 2009.

[5] You, S.H.N. Higuchi, S, Fujioka and T. Ueda; "Mechanism of Advective mixing in Non-Element Mixing in Non-Element Mixer", Kagaku Kogaku Ronbunshu, 35,570-597, 2009.

[6] Yuchao Zhao and Guangwen Chen and Quan Yuan, "Liquid-Liquid Two-Phase Flow Patterns in a Rectangular Microchannel" , Journal of Chemical Engineering of Japan,47, 213-224, 2014

[7] K.A. Triplett, S.M. Ghiaasiaan*, S.I. Abdel-Khalik, D.L. Sadowski, " Gas-liquid two-phase flow in microchannels. Part I: two-phase flow patterns", G. W. Woodru School of Mechanical Engineering, Georgia Institute of Technology, Atlanta, 1998.

[8] Ken-Ichiro Sotowa, "Fluid Behavior and Mass Transport Characteristics of Gas–Liquid and Liquid–Liquid Flows in Microchannels" Department of Chemical Science and Technology, the University of Tokushima, Journal of Chemical Engineering of Japan, Vol. 47, No. 3, pp. 213–224, 2014.

[9] Kashid M.N., A.Renken, L.Kiwi-Minsker, "Gas-liquid and liquid-liquid mass transfer in microstructured reactors", Chem.Eng. Sci. ,66,3876-389,2011.

[10] James R. Welty, Charles E. Wicks, Robert E. Wilson, Gregory L. Rorrer, "Fundamentals of Momentum, Heat, and Mass Transfer", 5th Edition, 2007.

[11] Kreutzer, M. T., F. Kapteijn, J. A. Moulijn and J. J. Jeiszwolf; "Multiphase Monolith Reactors: Chemical Reaction Engineering of Segmented Flow in Microchannels," *Chem. Eng. Sci.*, 60, 5895–5916 (2005)

[12] Rebrov, E. V.; "Two Phase Flow Regime in Microchannels," *eor. Found. Chem. Eng.*, 44, 355–367, 2010.

[13] Gupta, R., D. F. Fletcher and B. S. Haynes; "Taylor Flow in Micro-channels: A Review of Experimental and Computational Work," *J. Computational Multiphase Flow*, 2, 1–32, 2010.

[14] Sobieszuk, P., J. Aubin and R. Pohorecki; "Hydrodynamics and Mass Transfer in Gas–Liquid Flows in Microreactors," *Chem. Eng. Technol.*, **35**, 1346–1358, 2012.

[15] Yashihito Okubo, Taisuke Maki,Nobuaki Aoki, Teng Hong Khoo, Yoshikage Ohmukai,Kazuhiro Mae," Liquid-liquid extraction synthesis and separation by utilizing micro spaces". Chem. Eng. Sci., 63,4070-4077,2008.

[16] Nobuaki Aoki, Ryuichi Audo and Kazuhiro Mac, "Gas liquid –liquid slag Flow for improving Liquid-Liquid Extraction in Miniatured Channels", Ind.Eng.Chem.Res., 50, 4072-4677,2011.

[17] Akbar, M.K., Plummer, D.A., Ghiaasiaan, S.M., "On gas–liquid two-phase flow regimes in microchannels", International Journal of Multiphase Flow 29, 855–865,2003

[18] Usha Rao H., "Study of microscale fluid dynamics an input for micro reactor modeling and design "Ph.D., Thesis submitted to Osmania Universwity, 2012.

[19] Katherine León, Domingo Mery, Franco Pedreschi, Jorge León, "Color Measurement of L*a*b units from Digital Images", Elsevier, 2006.

[20] Anne-Laure Dessimoz, Laurent Cavinb, Albert Renkena, Lioubov Kiwi-Minskera, "Liquid–liquid two-phase flow patterns and mass transfer characteristics in rectangular glass microreactors", Elsevier, 2008.